

## Article

# Optimization of Solidification and Stabilization Efficiency of Heavy Metal Contaminated Sediment Based on Response Surface Methodology

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**Abstract:** Solidification and stabilization (S/S) by agents and stabilizers is an effective way to treat heavy metal-contaminated sediments. Optimization of curing condition is crucial to minimize the consumption of reagents on the base of effective S/S. In this work, the synergistic effects of cement and stabilizer on mechanical strength and leaching toxicity of contaminated sediments were investigated, and the S/S conditions were optimized using response surface methodology. On the basis of a single-factor test, multi-factor experiments were conducted to fit the relationship between the S/S effect of contaminated sediments and the amount of cement and stabilizer. The mechanism of stabilization was investigated by the results from the revised BCR method. The results indicate that the optimal curing conditions were 44.29% of cement content with 2.05% of trimercapto-s-triazine trisodium salt (TMT). After 28 days of curing, the compressive strength reached 2.07 MPa and the leaching concentrations of Cd, Cu, and Pb were 0.094 mg/L, 0.031 mg/L, and 0.173 mg/L, respectively, which met the requirement of in-situ resource recycling standard. The stability of heavy metals was significantly improved as a result of the removal of acid extractable fraction (15.58~69.92%) and an increase in the residual fraction (18.27~49.07%).

**Keywords:** TMT; unconfined compressive strength; leaching toxicity; revised BCR method; heavy metal fractionation



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## 1. Introduction

Sediment is an important part of aquatic ecosystems, which acts as both the source and the sink of pollutants [1]. Heavy metal contamination in sediments has become a global problem due to its high toxicity, persistence, and bioaccumulation behavior through the food chain [2,3]. A large number of rivers and lakes around the world exhibit varying degrees of heavy metal contamination, mainly caused by Cd, Cu, Pb, etc., with most showing compound contamination [4]. Thus, efficient treatment and disposal of heavy metal-contaminated sediment is crucial.

Solidification/stabilization treatment (S/S) of heavy metal-contaminated sediment has been widely applied in recent years. In this method, a curing agent, such as cement, is added to the contaminated sediment. It can not only prevent heavy metals from leaching into the environment, but also develop mechanical strength after curing, so that the treated sediment can be used as construction material [5–7]. The efficiency of S/S strongly depends on the amount of curing agent [8]. However, the capacity increment ratio and cost will significantly increase when stabilizing heavily contaminated sediments owing to the requirement of a high cement/water ratio to reduce the leaching of heavy metals [9]. What is more, production of most curing agents has a high carbon footprint, which is not acceptable for environmental purposes. The addition of a chemical stabilization reagent

may resolve this conflicting issue. It converts metals into low-toxicity and low-solubility forms through physical–chemical reactions, which can significantly reduce the mobility and bioavailability of heavy metals, thus avoiding excessive uptake and absorption of heavy metals by organisms [10]. Trimercapto-s-triazine trisodium salt (TMT) is an environmentally friendly agent that can chelate heavy metals with high thermal stability and much lower biological toxicity [11]. At present, S/S of heavy metal-contaminated sediments by chemical stabilizers and cement has been reported in many studies [12,13], but there are few systematic studies on the optimization of treatment conditions. Therefore, it is of great significance to investigate the feedstock ratio of compound curing agents to reduce the capacity increment ratio and treatment cost.

The sediments in the study were artificially prepared in a high concentration of heavy metal contamination. TMT and cement were selected as compound curing agents for S/S of contaminated sediment, and the effect was evaluated by examining the compressive strength, metal concentration in leachate, and metal fractionation after curing. Response surface methodology was used to optimize the feedstock ratio of the curing agent to minimize the use of cement and meanwhile ensure the resource utilization. This study may provide the important theoretical basis for the reduction, harmlessness, and reutilization of heavy metal-contaminated sediments.

## 2. Materials and Methods

### 2.1. Materials

The surface sediments in this experiment were from Fuxing Island Canal in Shanghai, China. The physio-chemical properties were water content 38.46%, pH 7.56, organic matter content 2.2%, and particle size distribution 5–75  $\mu\text{m}$ . The sediments were artificially contaminated by heavy metal salts  $\text{Cd}(\text{NO}_3)_2 \cdot 4\text{H}_2\text{O}$ ,  $\text{Cu}(\text{NO}_3)_2 \cdot 3\text{H}_2\text{O}$ , and  $\text{Pb}(\text{NO}_3)_2$ , according to the *Soil Environmental Quality Risk Control Standard for Soil Contamination of Agricultural Land (GB15618-2018)*. The sediment was well mixed and incubated for a month to make heavily contaminated sediments (Table 1). Normal Portland cement (PO.42.5) and a heavy metal-trapping agent (TMT-60,  $\text{Na}_3\text{C}_3\text{N}_3\text{S}_3 \cdot 9\text{H}_2\text{O}$ ) were used as the S/S reagent.

**Table 1.** Heavy metal content in sediment (mg/kg).

Heavy Metal	Cd	Pb	Cu
Raw sediments	0.280	21.50	68.25
Polluted sediments	120	1755	2941
Background values <sup>1</sup>	0.750	22.60	55.90

<sup>1</sup> Background values refer to the background values of heavy metals in sediments from Shanghai, this information was adapted from [14].

### 2.2. Experimental Design

Solidification specimens were made by referring to the *Standard of Geotechnical Test Method (GB/T 50123-1999)*. The water/cement ratio ranged from 10 to 60%. The prepared material was stirred with a mechanical stirrer and mixed evenly at a speed of 120 r/min. The material was loaded into a cylindrical mold (50 mm in height and 50 mm in diameter), and the mold was coated with a layer of Vaseline in advance). After 24 h, the mold was removed and placed in a standard curing box for curing (temperature  $20 \pm 1$  °C, humidity > 90%).

After curing for 7 days, 14 days, and 28 days, the unconfined compressive strength (UCS) test was carried out according to the *Standard of Geotechnical Test Method (GB/T 50123-1999)*. The machine used for the strength test was a DYE-300S integrated bending and compression machine. The specimen was placed on the machine and the machine was started at a speed of 0.10 kN/s, uniformly and continuously pressurizing the specimen until the specimen was destroyed, and the breaking load P at this time was recorded. Formula 1 was used to calculate the UCS of the specimen:

$$f = \frac{P}{A} \quad (1)$$

where  $f$  is the UCS (MPa),  $P$  is the breaking load (N), and  $A$  is the specimen-bearing area ( $\text{mm}^2$ ). The leaching toxicity test was carried out according to the *Solid Waste-Extraction Procedure for Leaching Toxicity—Acetic Acid Buffer Solution Method (HJ/T 300-2007)*. The internal broken sample destroyed by the compression test was ground and screened (9.5 mm) for later use.

Then, the extracting agent was prepared: 5 mL acetic acid was dissolved in 1000 mL pure water and titrated with acetic acid to  $\text{pH} = 2.64 \pm 0.05$ . A total of 2 g samples were taken, and 40 mL extracting agent was placed in a 50 mL centrifuge tube. The centrifuge tube was fastened to the cap and fixed on the rotating oscillation device at a rotational speed of  $(30 \pm 2)$  r/min, and the oscillation was performed at  $(23 \pm 2)$  °C for  $(18 \pm 2)$  h. After the oscillation, the sample was centrifuged for 10 min at a centrifuge speed of 5000 r/min, filtered with a needle filter of 0.45  $\mu\text{m}$ , and acidified with a drop of concentrated nitric acid to store for testing. The leaching concentration was tested by inductively coupled plasma mass spectrometry (ICP-MS, Nexion 300, Perkin Elmer, MA, USA). Three replicate samples were made in each experiment group.

TMT was directly incorporated into wet sediment at a dosage of 0.25~3.5%. The material was loaded into the mold and cured for 7 days after being fully mixed, and the leaching concentration was tested by ICP-MS. Three replicate samples were made in each experiment group.

### 2.3. Detection and Analysis

The UCS test was conducted by referring to the *Standard of Geotechnical Test Method (GB/T 50123-1999)*. The test of the leaching concentration of heavy metal was conducted in accordance with the *Solid Waste-Extraction Procedure for Leaching Toxicity—Acetic Acid Buffer Solution Method (HJ/T300—2007)*, in which an extraction buffer of acetic acid ( $\text{pH} = 2.64 \pm 0.05$ ) was used. The metal concentration in the leachate was analyzed by ICP-MS.

With the revised BCR sequential extraction method [15], heavy metals were fractionated as acid extractable, reducible, oxidizable, and residual fractionations. The procedures of the revised BCR extraction method are shown below. Acid extractable form (F1): 0.8 g sample was placed in a 50 mL plastic centrifuge tube. Then, 32 mL 0.11 mol/L  $\text{CH}_3\text{COOH}$  solution was added, shaken at room temperature for 16 h, and then centrifuged at 3000 rpm for 20 min. The supernatant was filtered through a 0.45 mm filter membrane, placed in a small square bottle, and stored in 2 drops of  $\text{HNO}_3$ . A total of 15 mL ultrapure water was added to clean the residue, and the supernatant was centrifuged and discarded. Reducible form (F2): 32 mL 0.5 mol/L  $\text{NH}_2\text{OH}\cdot\text{HCl}$  was added to the residue of F1 and then centrifuged at room temperature for 16 h. The other steps are the same as above. Oxidizable form (F3): 8 mL 8.8 mol/L  $\text{H}_2\text{O}_2$  (adjusted pH to 2–3 with 2 mol/L  $\text{HNO}_3$ ) was added to the residue of F2. The solution was intermittently oscillated in a centrifuge tube, digested at room temperature for 1 h, and then placed in an  $85 \pm 2$  °C water bath for 1 h until the solution was nearly dry. Then, 8 mL 8.8 mol/L  $\text{H}_2\text{O}_2$  was added, and the above steps were repeated. After completion, the sample was cooled, and then 32 mL 1 mol/L  $\text{CH}_3\text{COONH}_4$  was added to shake for 16 h at room temperature and centrifuged. The other steps are the same as above. Residual form (F4): The residue after F3 extraction was transferred to the PTFE crucible for digestion, and the digestion method was consistent with the method for measuring the total amount of heavy metals. The metal concentrations in each fraction were measured by ICP-MS.

### 2.4. Modeling by Response Surface Method

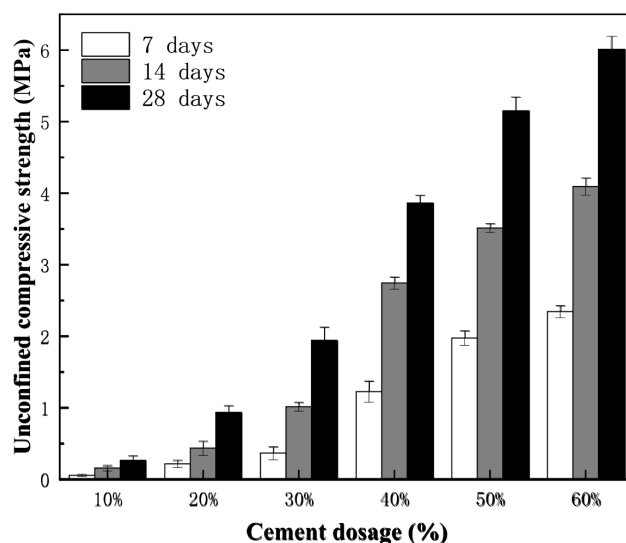
The central composite design (CCD) of response surface methodology was adopted to optimize the ratio of cement to TMT. With the testing parameters of all factors referring to the results of the single-factor test, the experimental results were fitted and optimized by

multiple stepwise regression technology with Design-expert 10.0 software, and the optimal experiment results were tested via a verification experiment.

### 3. Results

#### 3.1. The Effect of Cement on the Solidification/Stabilization of Sediment

Figure 1 shows that the UCS of the solidification specimens increased in response to the addition of cement and maintenance time. The UCS reached up to 6.01 MPa when 60% cement was added to the solidification specimens and cured for 28 days. According to the *Technical Standard for the Construction of Ecological Revetment by In-Situ Use of Dredged Sediments (DG/TJ 08-2331-2020)*, UCS of type II solidified soil should exceed 2 MPa at 28 days maintenance. This requirement was met when 40% of cement was added, as the UCS at 28 days maintenance reached 3.86 MPa.

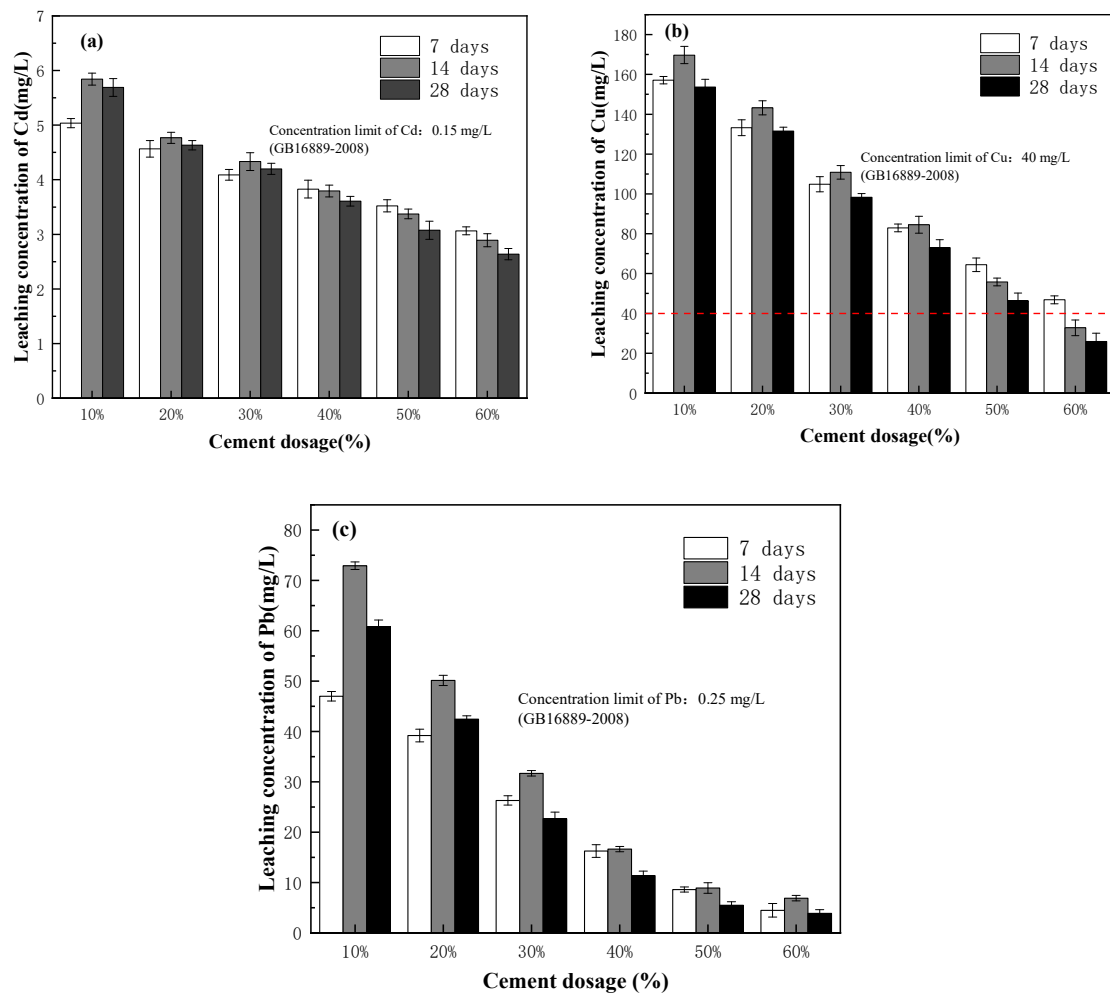


**Figure 1.** The change of unconfined compressive strength (MPa) of the specimens in response to the different cement dosage (%) and curing time (day).

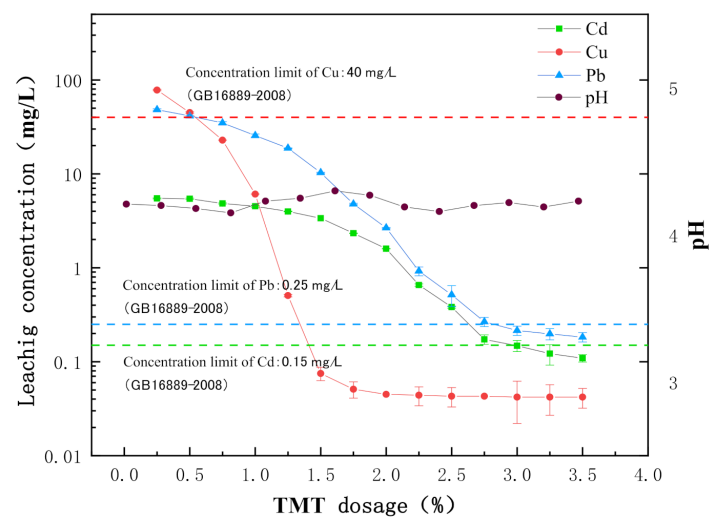
As shown in Figure 2, the leaching concentration of heavy metals of the solidification specimens decreased in response to the decrease in the water/cement ratio and the increase in curing time, which is attributed to the calcium silicate hydrate gel (C-S-H) formed by cement through the hydration reaction and coating the heavy metals in a gel system. More heavy metals were coated as the reacting time of the hydration reaction increased. Moreover, the system pH rose, resulting in the precipitation of heavy metals. Wang et al. studied the mechanism of cement-solidified  $\text{Cd}^{2+}$  and found that  $\text{Cd}^{2+}$  could be stabilized in the position between C-H-S layers through the formation of  $\text{Cd}(\text{OH})_2$  and  $\text{CdCO}_3$  precipitation, resulting in a decrease in the amount of leaching heavy metals [16]. Nevertheless, the results show that the Cd and Pb leaching concentration still exceeded the standard 15–240 times with the highest cement dosage (60%), cured for 14 days and 28 days. Only the Cu leaching concentration met the requirements of the *Pollution Control Standard for Domestic Waste Landfill (GB16889-2008)* of below 40 mg/L.

#### 3.2. Effect of TMT on Heavy Metal Stabilization

The stabilization effect of different dosages of TMT on the heavy metals in the sediment is shown in Figure 3. With the increasing amount of heavy metal trapping agent, the leaching concentration of heavy metals decreased continuously, and the leachate pH fluctuated slightly around 4.4. When the TMT content reached 3%, the leaching concentrations of Cd, Cu, and Pb were 0.148 mg/L, 0.042 mg/L, and 0.215 mg/L, respectively, which are below the limit in the *Pollution Control Standard for Domestic Waste Landfill (GB16889-2008)*.



**Figure 2.** The concentration of (a) Cd, (b) Cu, and (c) Pb in leachate under different cement dosages (%) and curing times (day). The red dash line represents the limit concentration of Cu in the *Pollution Control Standard for Domestic Waste Landfill (GB16889-2008)*.



**Figure 3.** The effect of TMT dosages (%) on the pH and heavy metal concentration in leachate from specimens after adding TMT to the sediment for 7 days. The dash lines represent the limit concentrations of Cu, Pb, and Cd in the *Pollution Control Standard for Domestic Waste Landfill (GB16889-2008)*.

### 3.3. Response Surface Test Results

The value range of each factor was determined based on the results of the single-factor experiment in Sections 2.1 and 2.2, and a two-factor and five-level experiment was designed to seek the optimal dosage of cement and TMT (Table 2).

**Table 2.** Test factors and horizontal design in the response surface model.

Coded Value	−1.414	−1	0	1	1.414
Cement dosages/A (%)	40.17	41.00	43.00	45.00	45.83
TMT dosages/B (%)	1.72	1.80	2.00	2.20	2.28

#### 3.3.1. Relationship between Test Conditions, Compressive Strength, and Leaching Concentration of Heavy Metals

The UCS and the leaching concentrations of metals were analyzed by response surface methodology. Stepwise regression simulation was performed to produce four models, which are shown in Table 3. The model's  $p$ -values were all  $<0.005$ , indicating that each model item reached a significance level. The  $p$ -values were all  $>0.05$ , indicating that the lack of fit items was not significant [17]. The correlation coefficients ( $R^2$ ) and the correction coefficients ( $R^2_{Adj}$ ) of the regression model were all above 0.92 and 0.85, respectively, except for the leaching concentration of Cu. The variability of most experimental data corresponded to the model.

**Table 3.** Model regression equation and variance analysis.

Regression Equation	$p$	PLOF	$R^2$	$R^2_{Adj}$
$UCS = -55.24 + 1.90A + 15.70B - 0.03AB - 0.02A^2 - 3.71B^2$	0.0001	0.9836	0.9588	0.9293
$C_{Cd} = 17.85 - 0.41A - 7.83B + 0.0069AB + 1.73B^2$	$<0.0001$	0.3486	0.9678	0.9447
$C_{Cu} = 0.27 - 0.0019A - 0.08B$	0.0002	0.789	0.8132	0.7759
$C_{Pb} = 42.97 - 1.56A - 7.74B + 0.12AB + 0.01A^2$	0.0009	0.1726	0.9231	0.8682

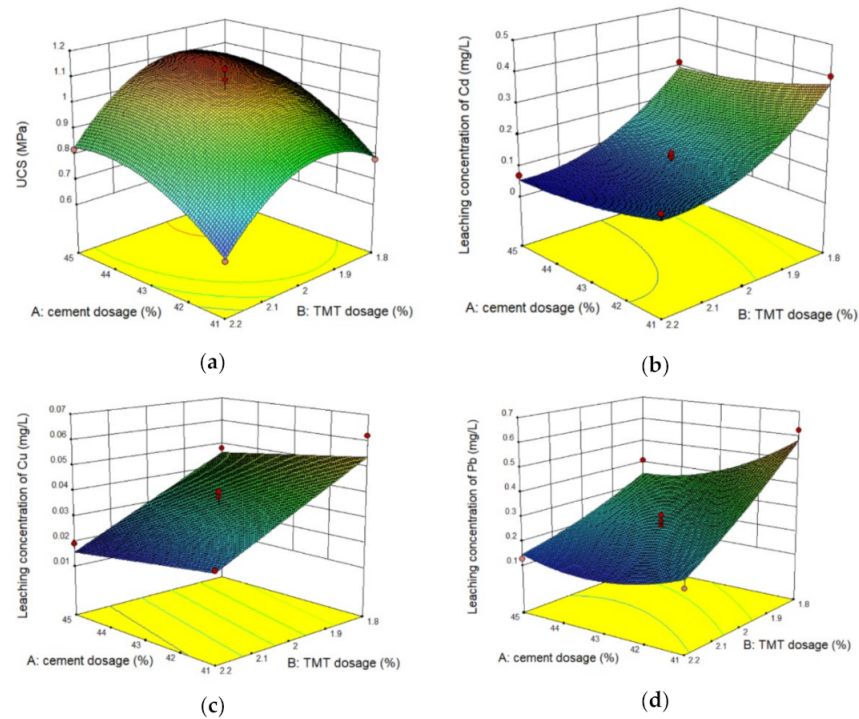
Note: A is for cement content; B is for TMT dosage;  $p$  is for probability of error; PLOF is for probability of lack of fit.  $R^2$  and  $R^2_{Adj}$  are the correlation coefficient and corrected coefficient, respectively.

#### 3.3.2. Interaction Analysis of Each Factor

Figure 4 shows the response surface plots between the UCS; the leaching concentrations of Cd, Cu, and Pb in solidified specimens; and the dosages of cement and TMT. The UCS increased significantly with the increase in cement dosage, whereas the mixing of TMT had an inhibitory effect on strength development. This is due to the fact that its complexes chelated with heavy metals were adsorbed on the surface of cement and clay particles, hindering and delaying the growth of hydration products, weakening the cementation inside the solidified body, and loosening its structure [18]. With the increase in cement dosage and reduction of TMT dosage, the three-dimensional plot of the response surface gradually showed an upward convex arc, indicating the obvious interaction between cement and TMT dosage, which suggests that the cement dosage can be reduced to achieve the maximum compressive strength under the premise of meeting the toxic leaching requirements.

As shown in Figure 4b, the leaching concentration only had a slight drop with the increase in cement dosage, which explains why the effect of the cement dosage on the leaching concentration of Cd was not significant, whereas the leaching concentration of Cd declined notably with the increase in TMT dosage. This indicates that in terms of the single-factor effect, the TMT dosage had the greatest effect on Cd leaching concentration. The interaction between two factors was especially evident when the cement dosage was below 43%.





**Figure 4.** Response surface of the interaction of cement dosage and TMT dosage on unconfined compressive strength (UCS, MPa) (a) and leaching concentrations of Cd (b), Cu (c), and Pb (d).

Figure 4c shows that the cement dosage had a negligible effect on the leaching concentration of Cu compared to the TMT dosage, which proves that the leaching concentration of Cu barely decreased with the increase in cement dosage, whereas the heavy metal trapping agent dosage had a significant effect on the leaching concentration of Cu. The leaching concentration of Cu decreased significantly with the increase in TMT dosage, which demonstrates that the TMT dosage was the dominant factor in the leaching concentration of Cu, which is consistent with the results of the significance analysis of the regression model. The plane shape of the response surface plot indicates that there was a remarkable effect of TMT on Cu stabilization, and there was almost no interaction between the two factors. Therefore, the AB term was excluded from the model.

Figure 4d shows that for the leaching concentration of Pb, the interaction of cement and TMT was obvious, especially when the cement dosage was greater than 43%. The gradual decrease in leaching concentration of Pb along with the increase in cement dosage was caused by the hydration reaction, in which  $\text{Pb}(\text{OH})_2$  crystals and  $\text{Pb}(\text{OH})_3^-$  ion complexes were formed by Pb through sequestration, which reduced the risk of Pb leaching [19]. The Pb leaching concentration decreased even more significantly with the increase in the heavy metal trapping agent, so TMT dosage is the main factor affecting the leaching concentration of Pb.

TMT has the best stabilizing effect on Cu, can complex almost all Cu, and has an excellent stabilizing effect on CD and Pb. It is the leading factor for the reduction of heavy metal leaching concentration, whereas cement mainly plays an auxiliary role in the stabilization of CD, Cu, and Pb. The cement hydration process forms an alkaline environment that makes the chelation products of heavy metals exist stably and promotes the stabilization of heavy metals. Moreover, the heavy metals wrapped by the hydration products can exist in a more stable and less mobile form [13]. Therefore, the leaching of heavy metals can be reduced and the specimen can be transformed from hazardous waste to cement-based materials to reduce and reuse solid wastes.

### 3.3.3. Optimal Ratio and Verification Experiment

Based on the previous analysis of the independent and interactive effects of the factors, the reaction conditions were optimized to find the optimal curing conditions, and the optimal ratio was obtained as 44.29% of the cement dosage and 2.05% of the heavy metal trapping agent. Then a verifying test was conducted with the purpose of exploring the S/S effects on heavy metal-contaminated sediment under the optimal ratio. The predictions of the model and experiment results are shown in Table 4, which shows that the predicted value was close to the test value, indicating that the regression model is reliable. Compared with the limits of the relevant standards, the UCS was higher than that of type II solidified soil required in the *Technical Standard for the Construction of Ecological Revetment by In-Situ Use of Dredged Sediments (DG/TJ 08-2331-2020)*, and the leaching concentrations of heavy metals were below the limits in the *Pollution Control Standard for Domestic Waste Landfill (GB-16889-2008)*, so the ratio could be used for the treatment and disposal of sediment in practical projects.

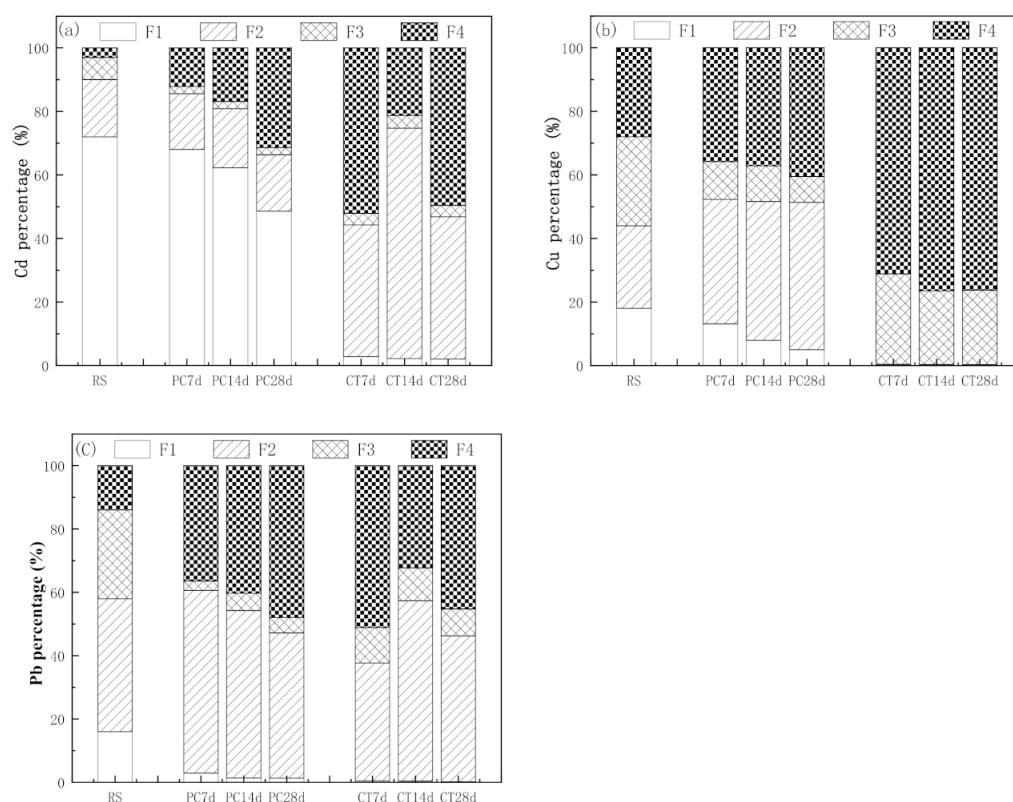
**Table 4.** Predicted and experimental results of unconfined compressive strength (Mpa) and heavy metal concentration in leachate of solidified and stabilized contaminated sediment treated under the optimal condition (44.29% cement and 2.05% TMT).

Items	UCS (Mpa)	Leaching Concentration (mg/L)		
		Cd	Cu	Pb
Predicted results	1.928	0.060	0.021	0.135
Experimental results	2.070	0.094	0.031	0.173
Target limit	2.000	0.15	40	0.250

### 3.4. Heavy Metal Fractionation

In order to further explore the transformation of heavy metal forms after solidification, the fractionations of heavy metals in sediment and curing blocks were classified into acid extractable fractionation (F1), reducible fractionation (F2), oxidative fractionation (F3), and residual fractionation (F4) by the revised BCR sequential extraction method, and the stability of heavy metals was improved in turn. Figure 5 shows the fractionation changes of Cd, Cu, and Pb in the raw sediments (RS); the Portland cement group (PC); and the cement-co-TMT group (CT) in response to the curing time. The Cd in the raw sediments mainly existed as high mobility F1 (72%), and the residual fraction occupied only 3%, with little difference between states of Cu and Pb. In the Portland cement group (PC), the acid extractable fraction decreased and the residual fraction (F4) increased with curing time, with F1 of Pb close to 0 and F4 close to 50%. Bao et al. also discovered that the amount of the residual fraction of Pb was highest in samples cured for 28 days [20]. This is because Pb is likely to form stable chelates with Fe/Mn oxides [21]. However, there was also a considerable amount of Cd in F1 (44%). In the cement-co-TMT group (CT), the mobile fractionation (F1) of Cd reduced to only 0.2%, whereas the stable fractionation (F4) increased to 50%, with the F1 of Cu and Pb close to 0 and the sum of the relative stable fractions (F3 and F4) up to 100%, meeting the effect of stabilization. This indicates that Cu and Pb was transformed into more stable fractions by TMT and existed as precipitates with the progress of the hydration reaction, which were sulfophilic and organophilic [22].





**Figure 5.** The change in the Cd (a), Cu (b), and Pb (c) fractionation in response to the curing time (days) and treatment. “7d, 14d and 28d” refers to the curing time are 7 days, 14 days and 28 days. RS is the raw sediment without treatment. PC refers to the S/S treatment with Portland cement only, whereas CT refers to the treatment with cement and TMT. F1 is the acid extractable fraction, F2 is the reducible fraction, F3 is the oxidizable fraction, and F4 is the residual fraction.

#### 4. Conclusions

When adding cement (40%, weight ratio) to solidify and stabilize heavy metal-contaminated sediments, UCS of 28 days can reach 3.86 MPa, meeting the requirement of the in-situ resource recycling of sediment, whereas the heavy metal leaching concentration is substandard according to the sanitary landfill requirement. The leaching concentrations of Cu, Cd, and Pb reached the sanitary landfill requirements with the addition of TMT (0.5%, 3.0%, and 3.0%, respectively, weight ratio) and the stabilization effect of TMT was Cu > Pb > Cd. Based on response surface methodology, the UCS and leaching concentrations of Cd, Cu, and Pb were fitted by stepwise regression simulation, and a model was constructed about the relationship between cement dosage and TMT dosage, which showed a good effect and can be further used to investigate the relationship between solidification condition, UCS, and leaching concentration of heavy metals, and the optimal annexing agent dosage conditions were obtained as 44.29% of cement and 2.05% of TMT.

The synergistic effect of cement and TMT significantly improved heavy metal stabilization compared to cement used alone. It considerably reduced the acid extractable and the reducible fractionation of heavy metals and increased the proportion of oxidizable and residual content, which reduces the mobility and potential ecological risk of heavy metals.

**Author Contributions:** Conceptualization, H.T. and Y.Y.; methodology, Y.Y. and M.L.; software, Y.Y. and M.L.; validation, Y.S.; formal analysis, Y.Y. and M.L.; investigation, Y.Y. and M.L.; resources, H.T.; data curation, M.L.; writing—original draft preparation, Y.Y.; writing—review and editing, Y.Y., H.T., L.M. and H.Z.; visualization, Y.Y. and H.G.; supervision, H.T., L.M. and H.Z.; project administration, H.T.; funding acquisition, H.T. All authors have read and agreed to the published version of the manuscript.

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**Conflicts of Interest:** The authors declare no conflict of interest.

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